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TOMOGRAPHIC RECONSTRUCTION OF INFRARED SPECTRA OF NONHOMOGENEOUS MEDIA: APPLICATIONS TO A FLAT FLAME BURNER

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13. ABSTRACT (*Maximum 200 words*)

We are using computed tomography, a radiological imaging technique, to obtain species profiles within a flame as a function of height above the burner surface and lateral position at that height. This information is extracted from parallel line-of-sight absorbance data using a technique known as Abel inversion. This process yields two dimensional "slices" of the flame. At any point within each slice the infrared spectrum of the species present may be obtained. Abel inversion is a special application of computerized tomography which may be applied only to systems possessing axial symmetry.

We have used this technique to evaluate line-of-sight infrared spectra of a low pressure (< 100 torr) premixed methane/nitrous oxide burner flame. The flame is supported on a water-cooled, stainless steel cylindrical frit which sits inside a low pressure chamber. Parallel line-of-sight infrared spectra through the low pressure chamber are inverted to give the radial dependence of infrared absorption at each wavelength of the spectrum. This data is used to evaluate the performance of the low pressure burner and to discriminate against "cold" gases which may obscure absorbances of trace species within the flame.

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1. INTRODUCTION

The study of low pressure flames has significance to the Army because these flames may provide information regarding the initial decomposition mechanisms of nitramine-based propellants (Schoeder 1980). In general, the flames under study are mixtures of a nitrogen-containing oxidizer and a conventional fuel. In the work reported here, a stoichiometric methane/nitrous oxide gas mixture is used because of the ease of working with this flame.

In a low pressure environment, the structure of a flame is expanded. Flame regions which hold the most interest for chemists and physicists are those in which chemical bonds are being broken and formed. These regions are called "reaction zones." At normal (atmospheric) pressures these reaction zones (i.e., preheat, primary, and secondary) are small compared to the overall flame dimensions (Fifer 1984). A low pressure environment causes the entire flame to become more diffuse, expanding the reaction zones and providing a larger region for study.

Although flames have been studied extensively since the middle of the 19th century (Gaydon 1974), there are experimental difficulties which have yet to be entirely overcome. Chief among these are the determination of temperature and the quantification of species within the flame. Ideally, any method of probing a flame should be quantitative and nonintrusive. Infrared spectroscopy is an excellent tool for such studies, but has the disadvantage that it is a line-of-sight technique, and therefore its value as a diagnostic tool is degraded when the beam must traverse a nonhomogeneous medium.

In this study, we report results from an analysis of a low pressure stoichiometric methane/nitrous oxide flowing mixture prior to and during combustion using Fourier transform infrared (FT-IR) spectroscopy. As mentioned above, the main disadvantage to using line-of-sight absorption techniques is that the signal at the detector is determined by species along the entire beam path. In low pressure flame work, large temperature and density gradients along the beam path make it difficult to determine which features in the spectrum are from species within the flame zone, and which are due to exhaust gases, or to species within interfacial regions, or species in the beam path outside of the low pressure chamber.

Several methods have been employed to discriminate against cold gas absorptions in line-of-sight spectra of burner flames. A shroud gas is often used to provide an entertainment of combustion species within the cylinder defined by the burner frit circumference. However, the probe beam still must traverse

regions of greatly varying density and temperature. Sapphire tipped "eyeballs" may be inserted into the chamber near the flame and used to physically exclude the beam from passing through the cold gas region, but this technique has the potential to perturb the flame. Another method of cold gas discrimination is to observe transitions from states which are only populated at elevated temperatures (Ouyang and Varghese 1991). This method still is limited by the fact that the technique is a line-of-sight method, and that the observed absorbance is still integrated along a path through all temperature and density gradients.

Recently, Best et al. (1991) have published a paper describing the history and use of tomography to determine species concentrations and temperatures within an atmospheric pressure diffusion flame, and to report the first use of tomography to analyze infrared spectra. We have applied a similar technique to the analysis of line of sight spectra of subatmospheric pressure flow mixtures prior to and during combustion. We have also used the technique to reconstruct full local spectra within the cylinder defined by our burner frit circumference.

2. BACKGROUND

Classical tomography assumes any object to be composed of a series of slices that have been stacked upon each other to form the object. The objective of classical tomography is to be able to look inside an object and see any one of those slices, with the view being unobscured by the slices in front of or behind the slice of interest. Typically, in classical tomography, only the layer of interest is in focus, while other layers are blurred or do not appear at all. As employed in radiology, classical tomography moves the source (usually emitting x-rays) and the detector in such a way that only a point in the plane of interest is in focus. This is usually accomplished by making the point of interest in the plane of interest the fulcrum about which the source-detector pair is moved. A reconstruction of the slice or plane is accomplished by taking a series of projections through different points in the plane of interest (Barrett and Swindell 1981). Computed tomography differs from classical tomography in that all projections in computed tomography are taken through the plane of interest, with the projections restricted to lie in that plane. The picture of the plane of interest is then reconstructed using computer techniques.

3. EXPERIMENTAL

The low pressure burner apparatus is shown in Figure 1. The flame is supported on a water-cooled, stainless steel fritted burner (McKenna Industries, Inc). The premixed fuel/oxidizer mixture is regulated

by an MKS Instruments Model No. 147 flow controller and flows into the bottom of the burner into a small volume directly below the porous frit. The stainless steel burner frit (6-cm diameter) is surrounded by a porous bronze annulus, through which a shroud gas may be flowed. The burner is mounted on a motorized stage, which provides horizontal translation. This stage is, in turn, mounted on a rotatable vertical positioning device, thus providing the capability of positioning the burner at any point within a Cartesian coordinate system defined by the ranges of the positioning devices. This whole assembly sits inside an evacuable chamber which has been equipped with LiF windows. A combined fuel/oxidizer flow rate of 2 L/min is typical to support a 30-torr flame.

Figure 2 shows the experimental apparatus used to probe the flame. The output beam from a Mattson Instruments Galaxy Series FT-IR spectrometer is passed through the center of the low pressure chamber. The output beam of the FT-IR spectrometer is brought to a focus at the center of the burner chamber (unapertured beam waist of ~1 cm). Detection of the interferogram output of the FT-IR spectrometer is by a liquid nitrogen-cooled HgCdTE detector. All spectra are obtained using 200 scans at 4-cm^{-1} resolution. Maximum apertured probe beam diameter is 4.7 mm. The output of the HgCdTe detector is transformed using triangular apodization. No zero filling is employed prior to the transformation of the interferogram. Also, no adjustment was made to account for any nonlinearity of the detector when exposed to the high thermal signal from the burner flame, since the thermal emission of the flame differs as different vertical positions in the flame are imaged. Normally, this introduces a DC offset to the signal reported by the detector. However, since only the probe beam is modulated, signal detection is not noticeably affected.

Figure 3 shows an example of the probe beam path for the experiments reported here. Assuming that the beam passes through a nonabsorbing medium outside of the burner chamber, there are three distinct regions of temperature and density within the burner chamber (exhaust, shroud, burner). Proceeding from the edge of the chamber to the burner center, the gas temperature increases and the gas density decreases. This means that a volume of exhaust gas outside the burner region (lower temperature, higher density) will absorb more strongly than the same volume of exhaust gas in the burner region (higher temperature, lower density). We believe the presence of exhaust gases accounts for the lower than expected temperatures often calculated from line-of-sight spectra (McNesby and Fifer 1991).

4. METHOD OF DATA REDUCTION

In the tomographic analysis employed here, the column of premixed gas and flame (if present) above the burner frit is assumed to have axial symmetry. Referring to Figure 4, let f(p) be the line-of-sight absorbance along a path normal to a radius vector r, where p is the distance of the normal to the path to the burner axis. The line-of-sight absorbance f(p) at a particular frequency is given by

$$f(p) = 2\int_p^1 g(r)rdr/(r^2 - p^2) , \tag{1}$$

where g(r) is the radial dependance of the absorbance at a particular frequency. Figure 5 shows f(p) as a function of p for a constant g(r) and for a triangular g(r). Equation 1 is one half of an Abel inversion pair (Cormack 1963), the other equation being

$$g(r) = -d/dr\{\pi/r\int_r^1 f(p)dp/[p(p^2 - r^2)]\} . \tag{2}$$

Figure 6 shows the result of inverting the synthetic data for the constant g(r) shown in Figure 5, with and without line-of-sight data through regions where the absorbance is zero, for 23 and 20 evenly spaced data points, respectively. The g(r) retrieved by the inversion agrees well with that input into the original calculation. The "ringing" observed in the region of transition between constant and zero absorbance decreases as the number of evenly spaced data points increases. The inversion program was kindly provided by Prof. Philip Varghese of the University of Texas (Deutsch and Beniaminy 1983).

5. RESULTS AND DISCUSSION FROM TOMOGRAPHIC ANALYSIS OF EXPERIMENTAL DATA

The tomographic technique just described is first applied here to a noncombusting stoichiometric CH_4/N_2O mixture, with an Argon shroud, flowing at 4.6 torr. Flow rates for the methane-nitrous oxide mixture were 0.35 and 1.3 L/min, respectively. Ar flow was 5.0 L/min. Because the technique should discriminate against all absorbances except those which change as beam position through the burner

changes, single-beam spectra are used in the analysis. Figure 7 shows a three-dimensional representation of line-of-sight single-beam spectra through the flowing $Ar/CH_4/N_2O$ mixture. These spectra represent the convolution of the source output and the detector response curves superimposed upon the absorptions due to gases in the beam path. The probe beam is centered at the lowest unobscured position relative to the burner surface (beam center 2.3 mm above burner surface). The absorption caused by the asymmetric stretch of CO_2 (in the beam path outside of the chamber) dominates the spectrum. Features due to the C-H stretch in methane (3,020 cm^{-1}) and the asymmetric stretch in N_2O (2,223 cm^{-1}) are much less discernable. In addition, there is no appreciable diminishment in absorption due to CH_4 or N_2O for line-of-sight spectra as the edge of the burner frit (at 30 mm off-axis distance) is approached by the probe beam. Figure 8 shows the spectrum for $r = 0$ (i.e., at the burner center) reconstructed using Abel inversion from the data in Figure 7. For this reconstruction, the absorbance due to the asymmetric stretch of CO_2 has been almost completely removed. An absorption is shown as a negative deviation since gases present in the beam path remove intensity from the beam. The increase in noise relative to the original spectra is caused by the inversion process. Usually, some form of data smoothing (Best et al. 1991) (and accompanied decrease in spectral resolution) is performed prior to inversion of the data to minimize the noise in the transformed spectra. No smoothing of the data was performed for the spectra reported here.

Figure 9 shows a three-dimensional reconstruction of the radial dependence of the infrared spectrum of the flowing gas mixture obtained from the spectra presented in Figure 7. At the right (low frequency) side of this figure the N_2O absorption (at 2,223 cm^{-1}) is seen to diminish (become less positive) as the edge of the burner frit is approached. Similar behavior is seen at 3,020 cm^{-1} for the C-H stretch of CH_4. The unusual behavior of the absorption due to the asymmetric stretch of CO_2 is believed to be due to atmospheric fluctuations in CO_2 during the course of a given experiment, which is accentuated because the beam travels approximately 1 meter through open air between the spectrometer and the low pressure chamber. Figure 10 shows reconstructed spectra of the species present at the burner center as a function of height above the burner surface. These spectra were reconstructed from the spectra shown in Figure 7. Intensities have been normalized to 0 at 2,000 cm^{-1} to aid in comparing the spectra to each other. In general, it appears that the concentrations of methane and nitrous oxide remain nearly constant at the burner center up to 23 mm above the burner surface.

Figure 11 shows a three-dimensional representation of line-of-sight absorbance spectra taken through a stoichiometric methane/nitrous oxide flame at 40-torr total pressure. No shroud gas is used. The 4.7-mm diameter probe beam is centered 6 mm above the burner surface. Spectra are collected at 4-cm^{-1}

resolution. The spectral region shown is that corresponding to the asymmetric stretch region of CO_2 and N_2O. In this figure, the CO_2 absorbance is seen to obliterate all other features. Figure 12 shows the data from Figure 11 after inversion. The CO_2 absorbance has now almost completely disappeared and, instead, an absorbance due to N_2O (2,223 cm^{-1}) is the most prominent feature in the spectra. The broad profile of the N_2O absorption is believed to be caused by the temperature gradient which occurs along the distance defined by the probe beam diameter (4.7 mm). This reconstruction indicates that the CO_2 absorbance in Figure 11 is due to gas outside the cylindrical region proscribed by the burner frit circumference. No absorbance due to CH_4 at 3,020 cm^{-1} was observed in either the raw or transformed spectra. The gradual sloping of the absorbances as off-axis distance (p) increases in the inverted spectra in Figure 12, may in part be caused by the limited number (8) of data points.

6. CONCLUSIONS

We believe that tomographic analysis of line-of-sight spectra through inhomogeneous media will become a standard analytical technique. While many experimental and computational difficulties remain, the degree of sophistication already employed in radiological imaging will undoubtedly accelerate development of tomography as a spectroscopic technique. We are engaged in an ongoing research project which is applying tomographic analysis to laser spectroscopy and Fourier transform spectroscopy with the aim of being able to spatially quantify all infrared active species within the burner flame. At present, our immediate goal is to increase the number of projections obtainable with the Fourier transform spectrometer, since the quality of the reconstruction increases with the number of projections. We believe this will become a valuable tool in combustion spectroscopy. We are also developing an application of the technique to FT-IR microscopy.

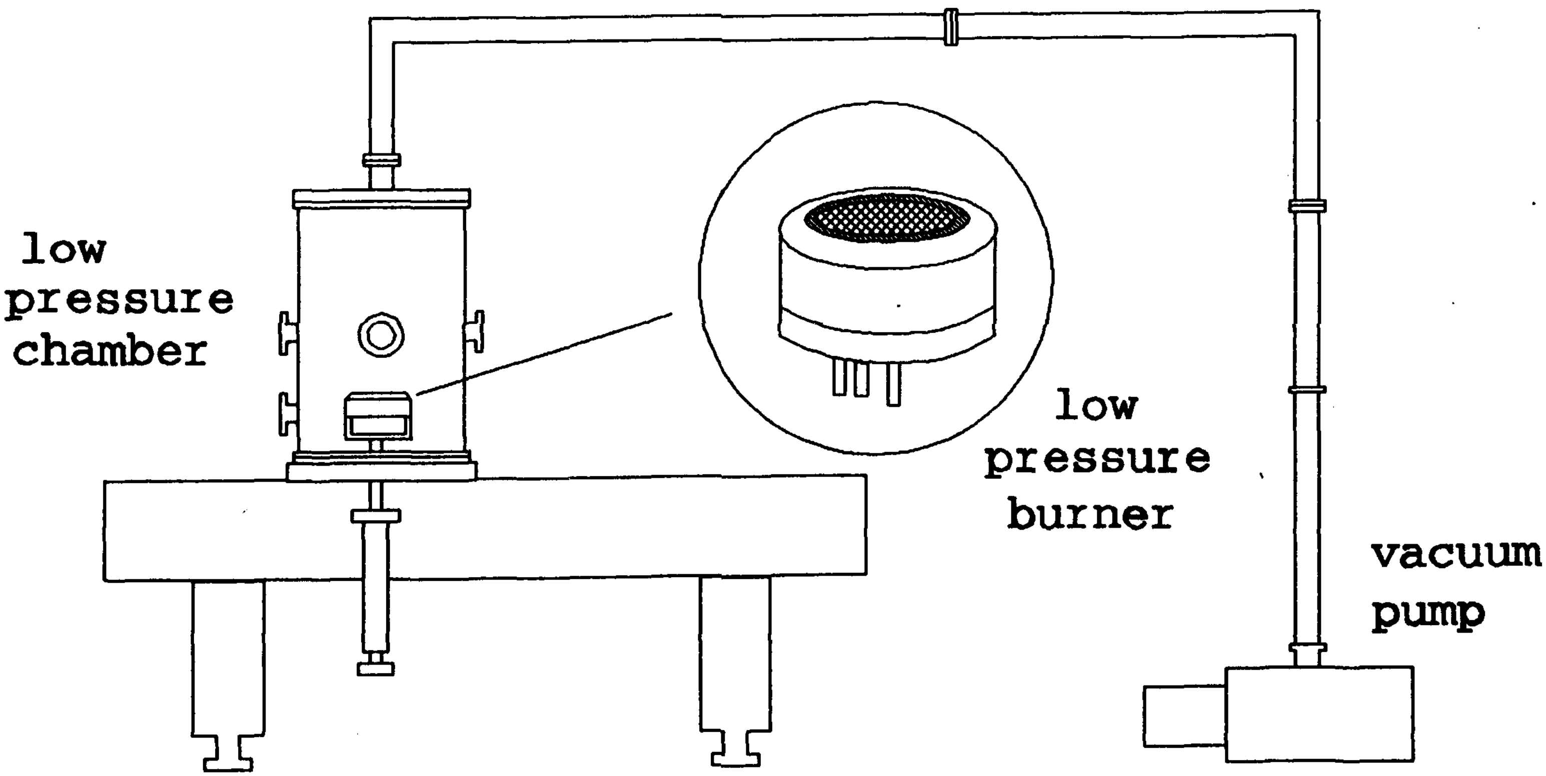

Figure 1. The Low Pressure Burner Apparatus Used in the Experiments, Showing the Burner Inside Evacuable Housing.

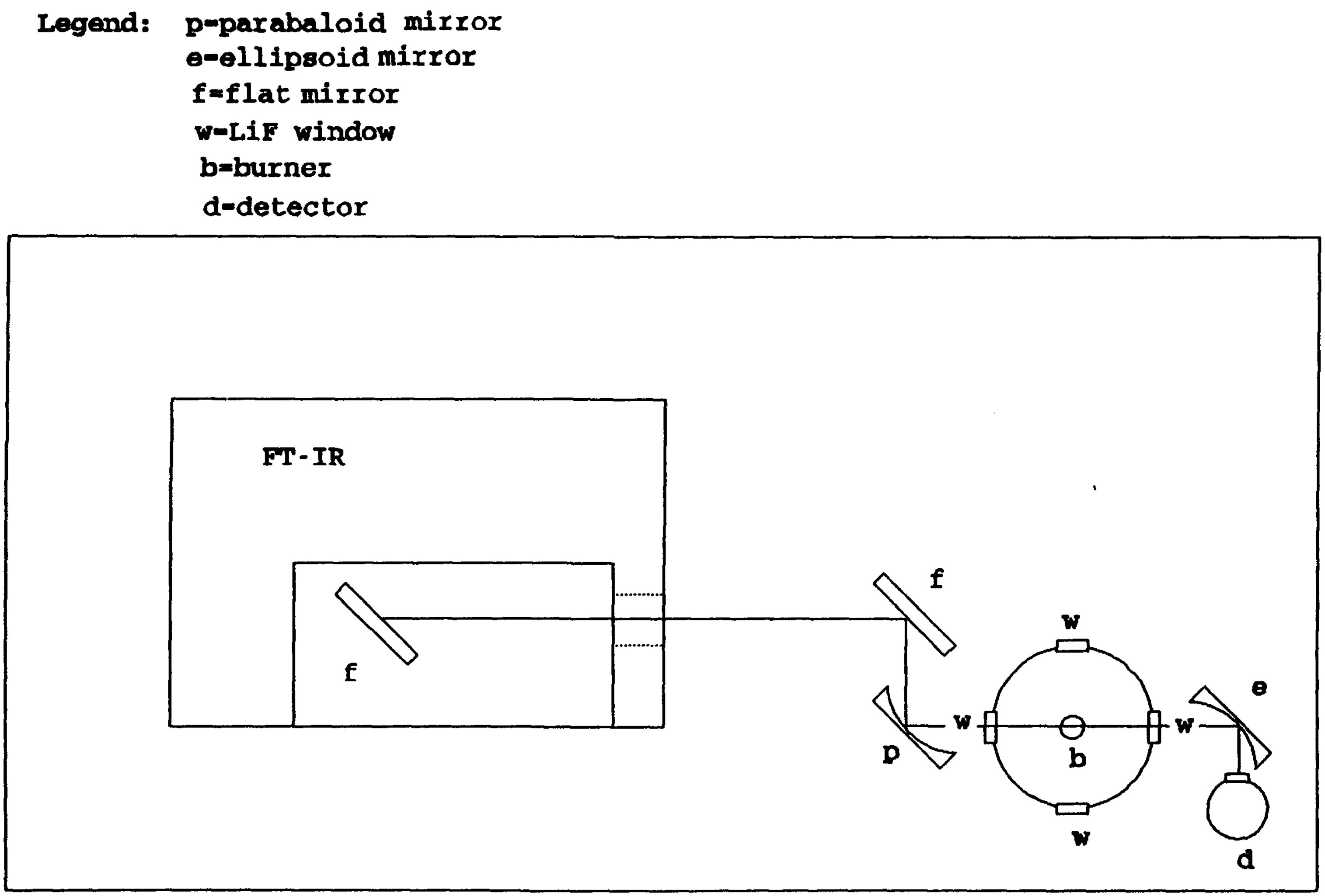

Figure 2. The Experimental Apparatus Used to Probe Low Pressure Flames.

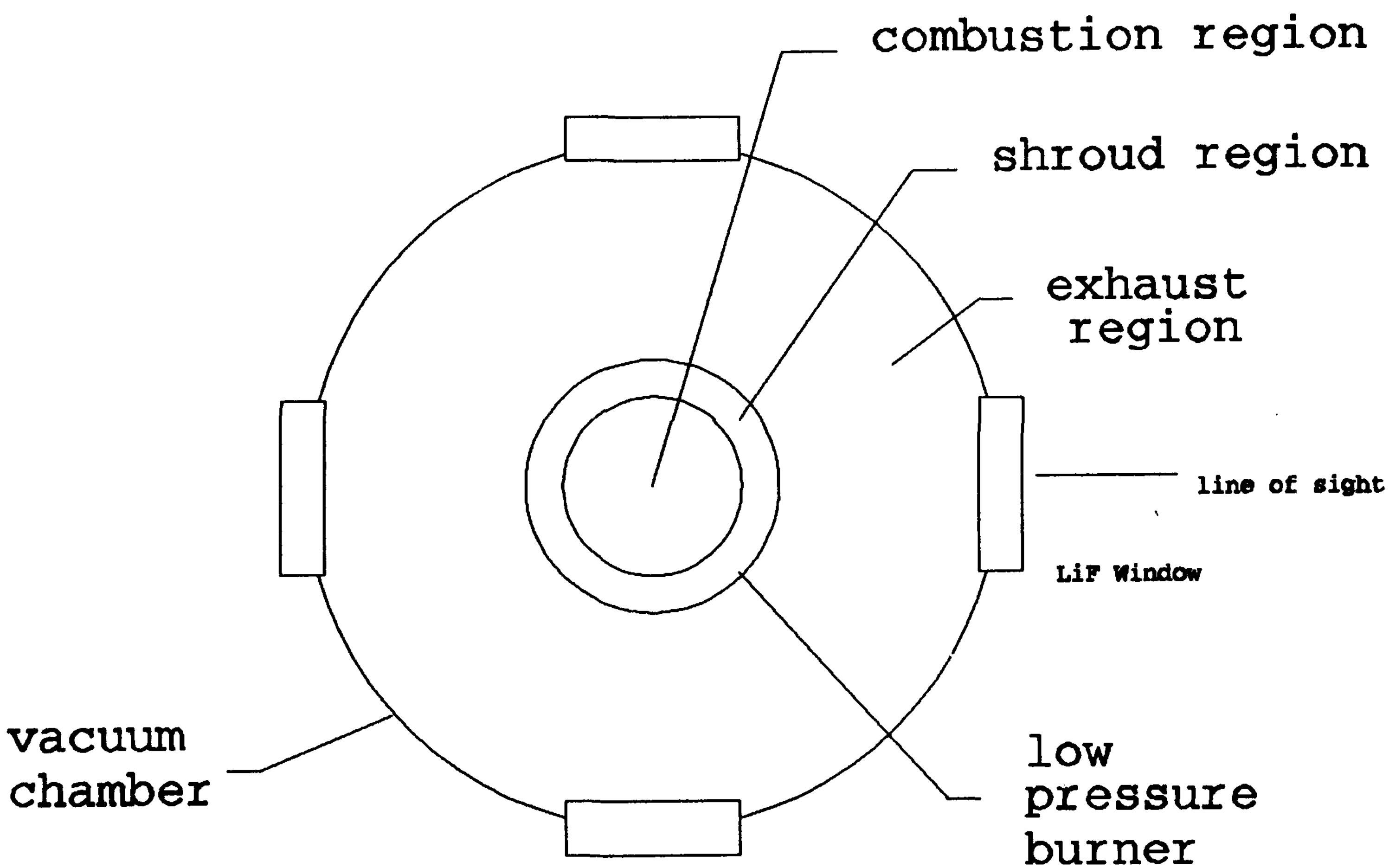

Figure 3. <u>The Different Regions Traversed by the Probe Beam in Infrared Absorption Measurements of Low Pressure Flames.</u>

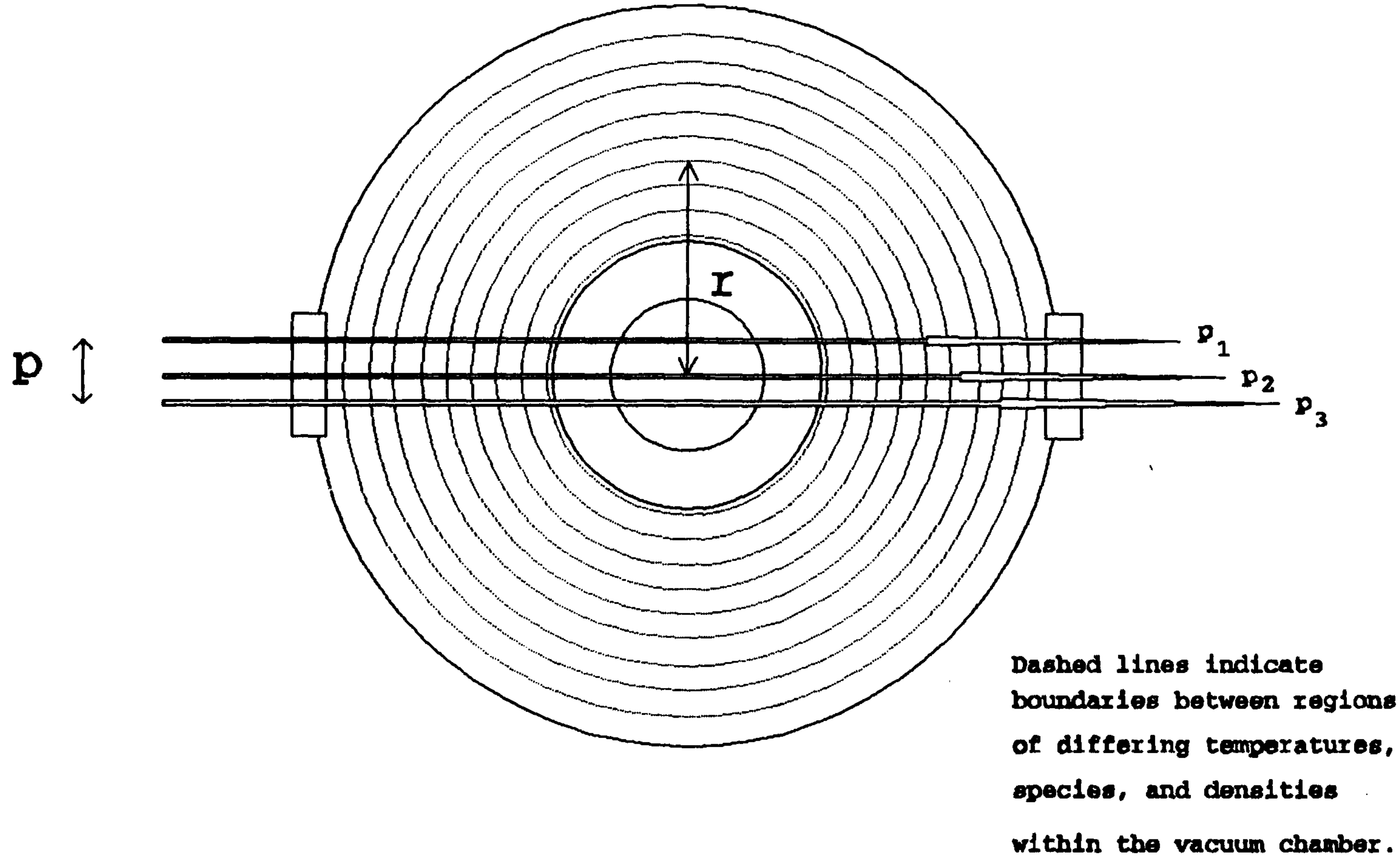

Figure 4. A Description of the Line-of-Sight Measurements Used for Tomographic Analysis of Data.

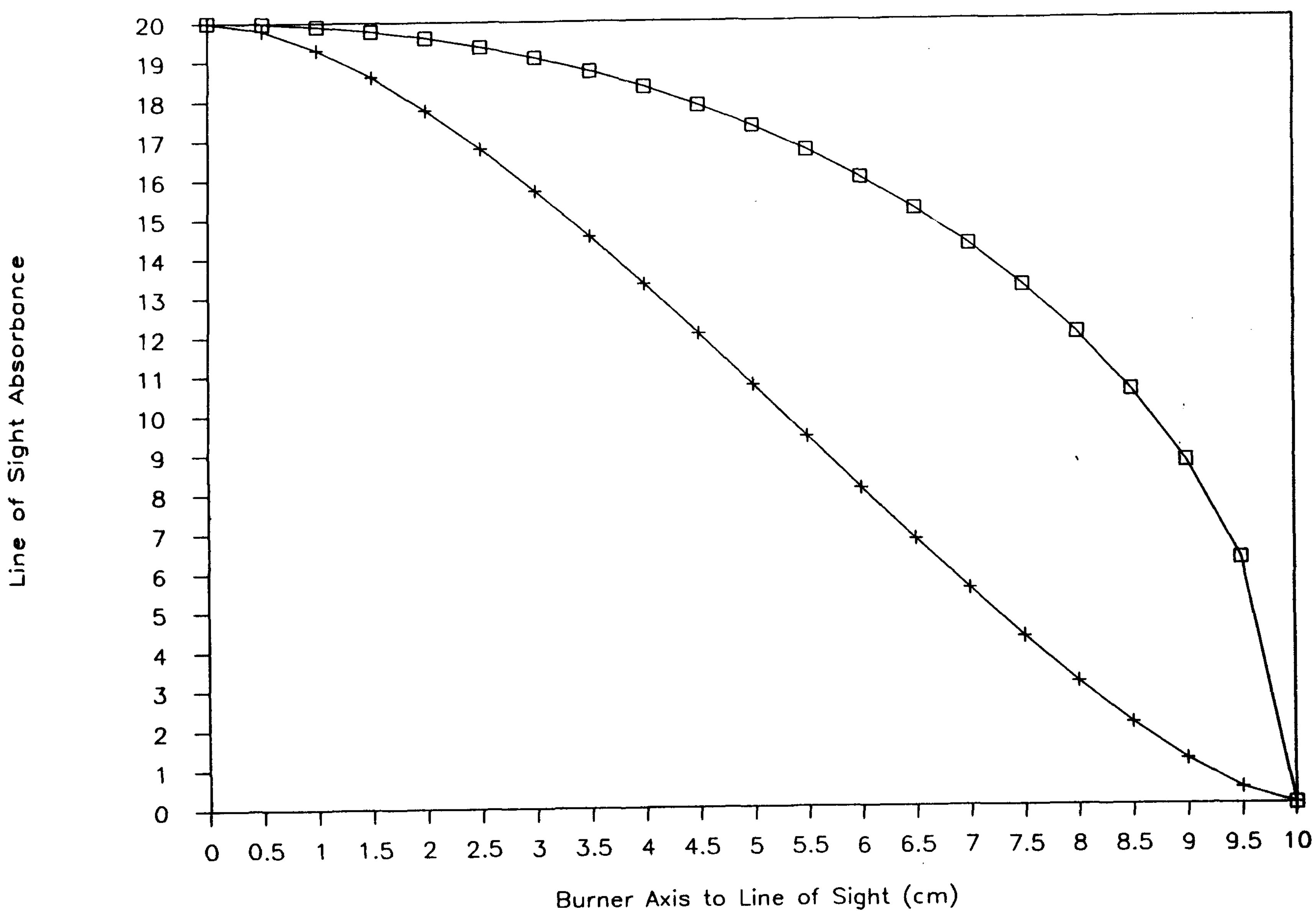

Figure 5. Synthetic Line-of-Sight Absorbances as a Function of Off-Axis Position for Constant and Varying $g(r)$. Plusses Denote $g(r) = A_o(1 - r/D)$. Squares Denote $g(r) = A_o$. D is the Burner Diameter.

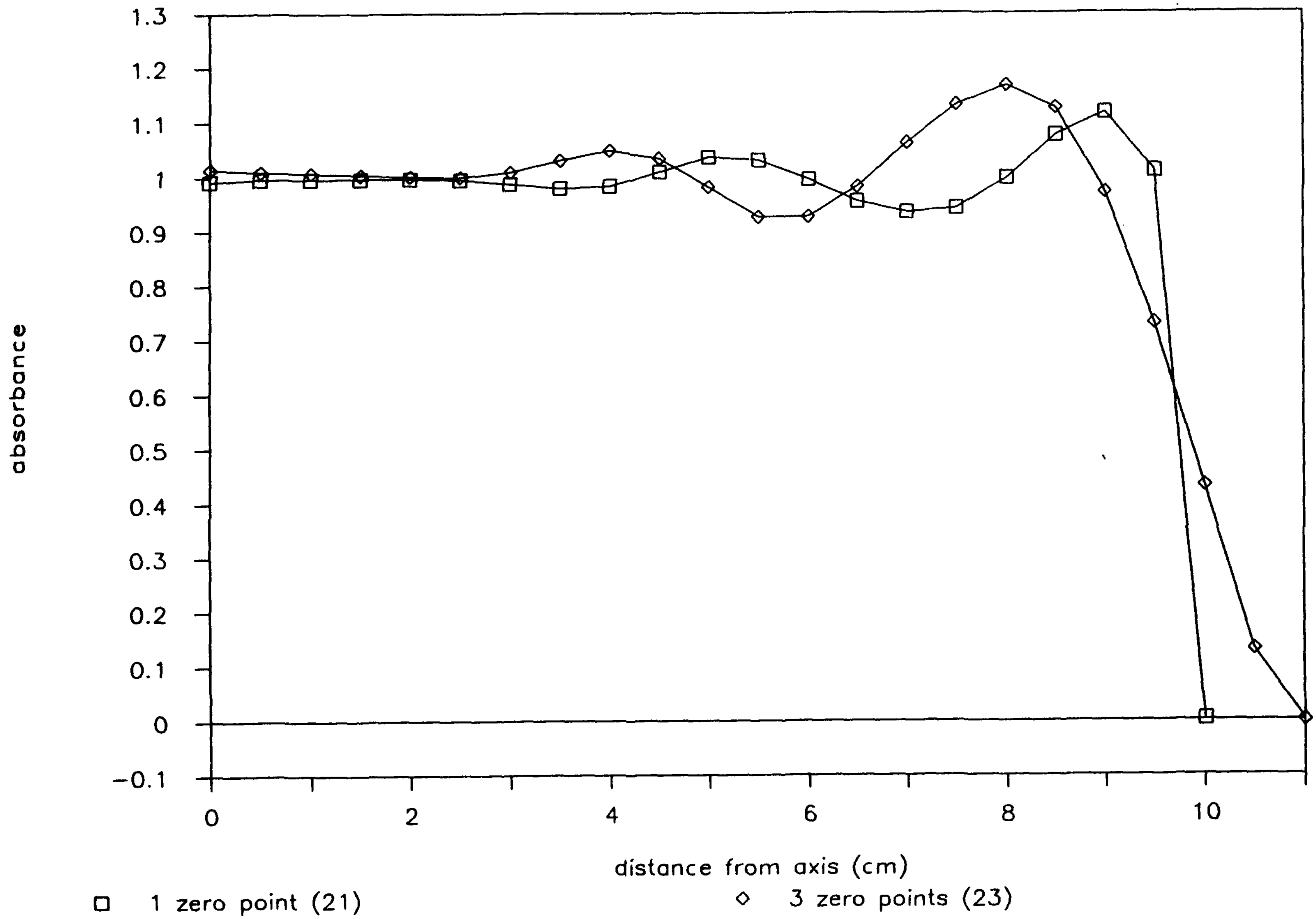

Figure 6. Inverted Synthetic Data for Constant g(r) Using 21 and 23 Data Points.

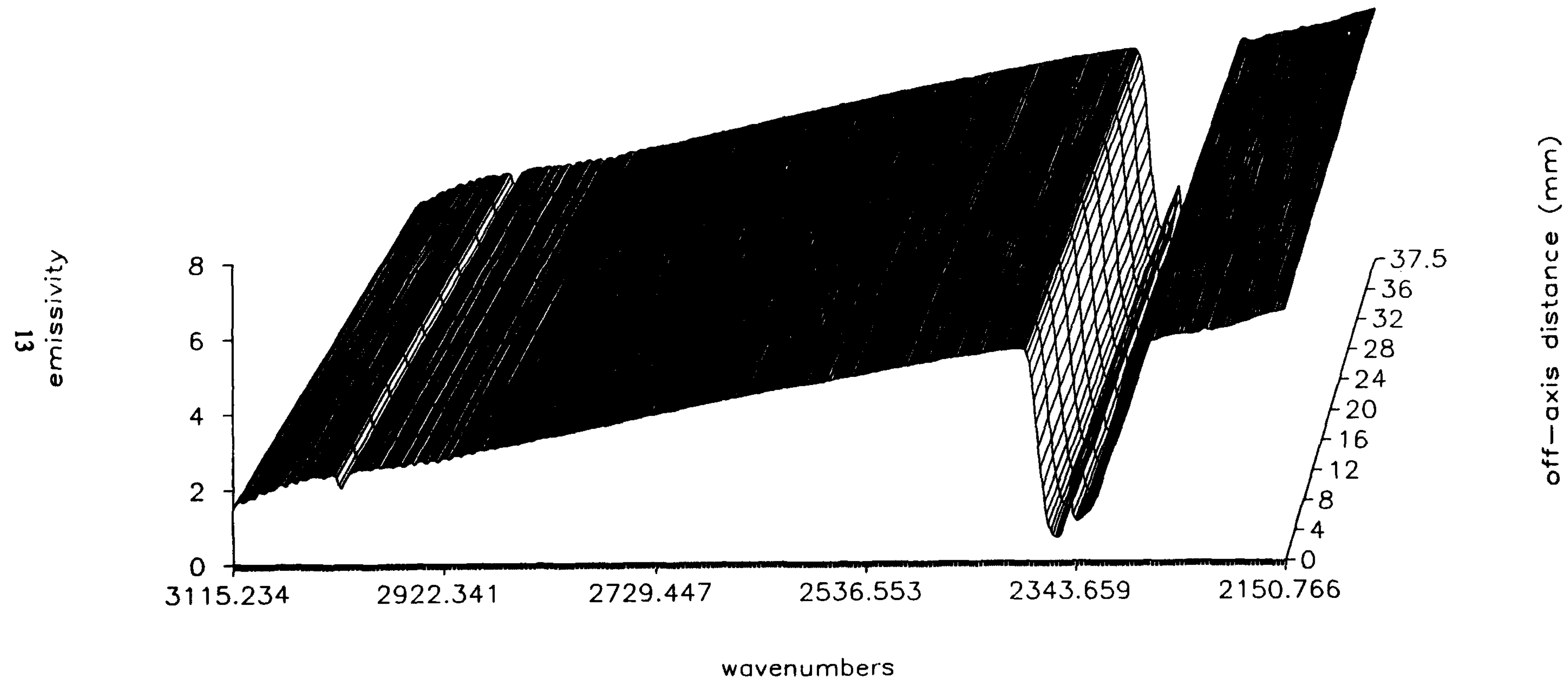

Figure 7. Line-of-Sight Beam Spectra Through a Flowing $Ar/CH_4/N_2O$ Mixture at 4.5 torr.

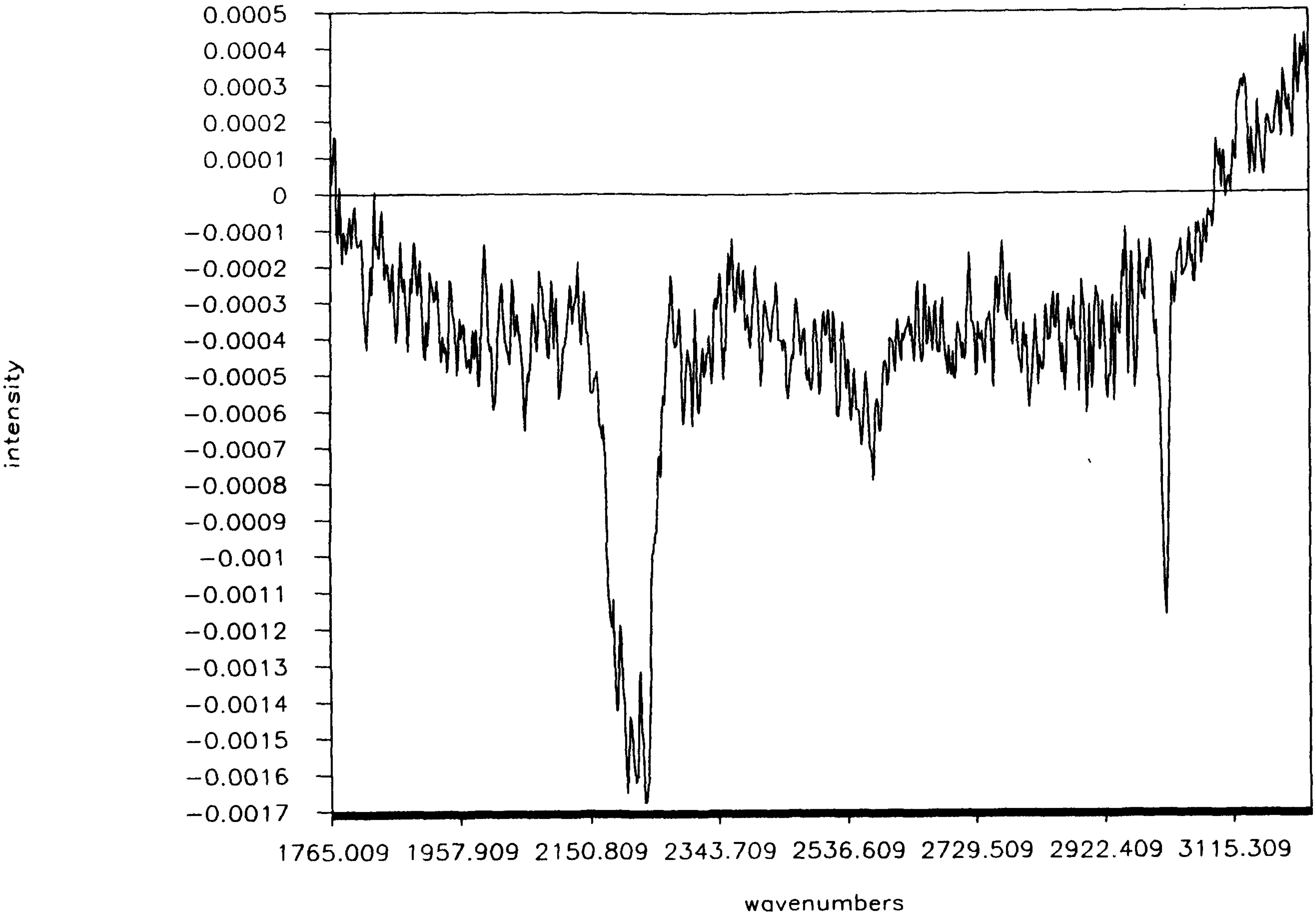

Figure 8. <u>Infrared Spectrum at the Burner Center (r = 0) Reconstructed From the Data Shown in Figure 7.</u>

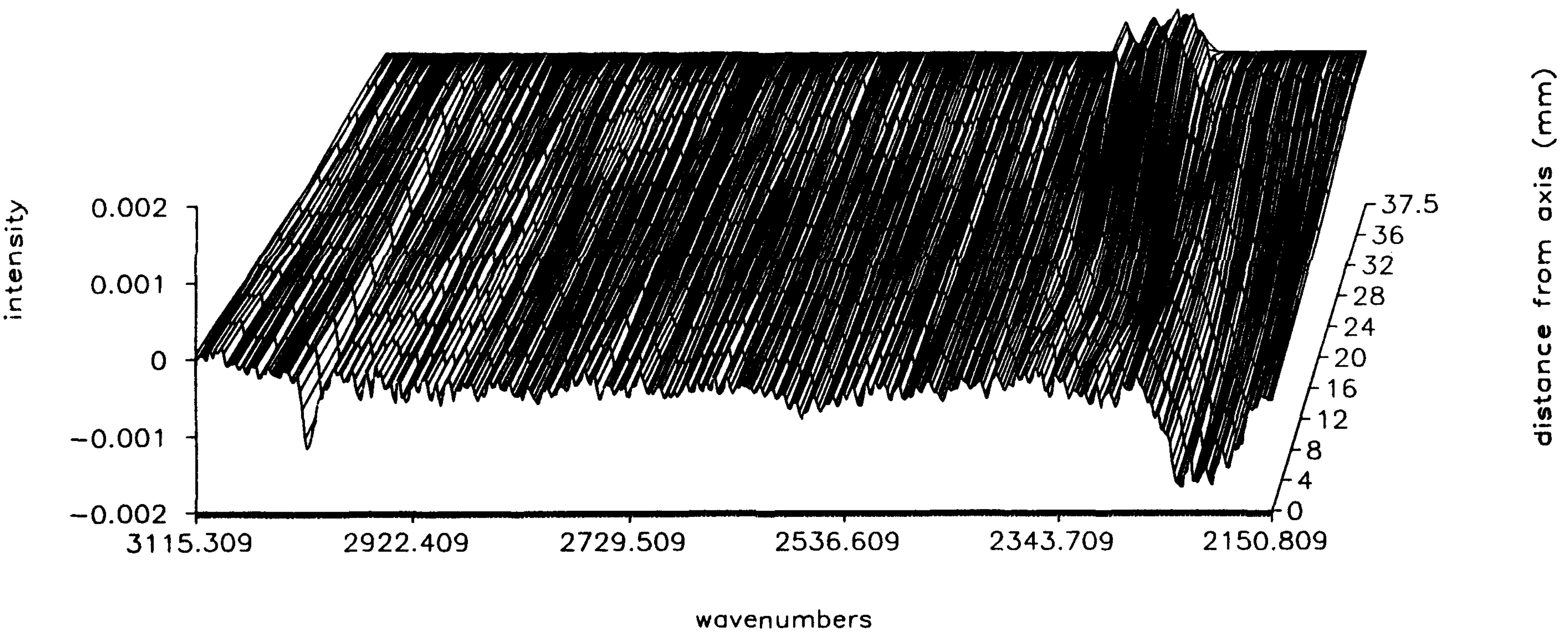

Figure 9. Radial Dependence of the Infrared Spectrum of the Flowing $Ar/CH_4/N_2O$ Mixture Reconstructed From the Data Shown in Figure 7.

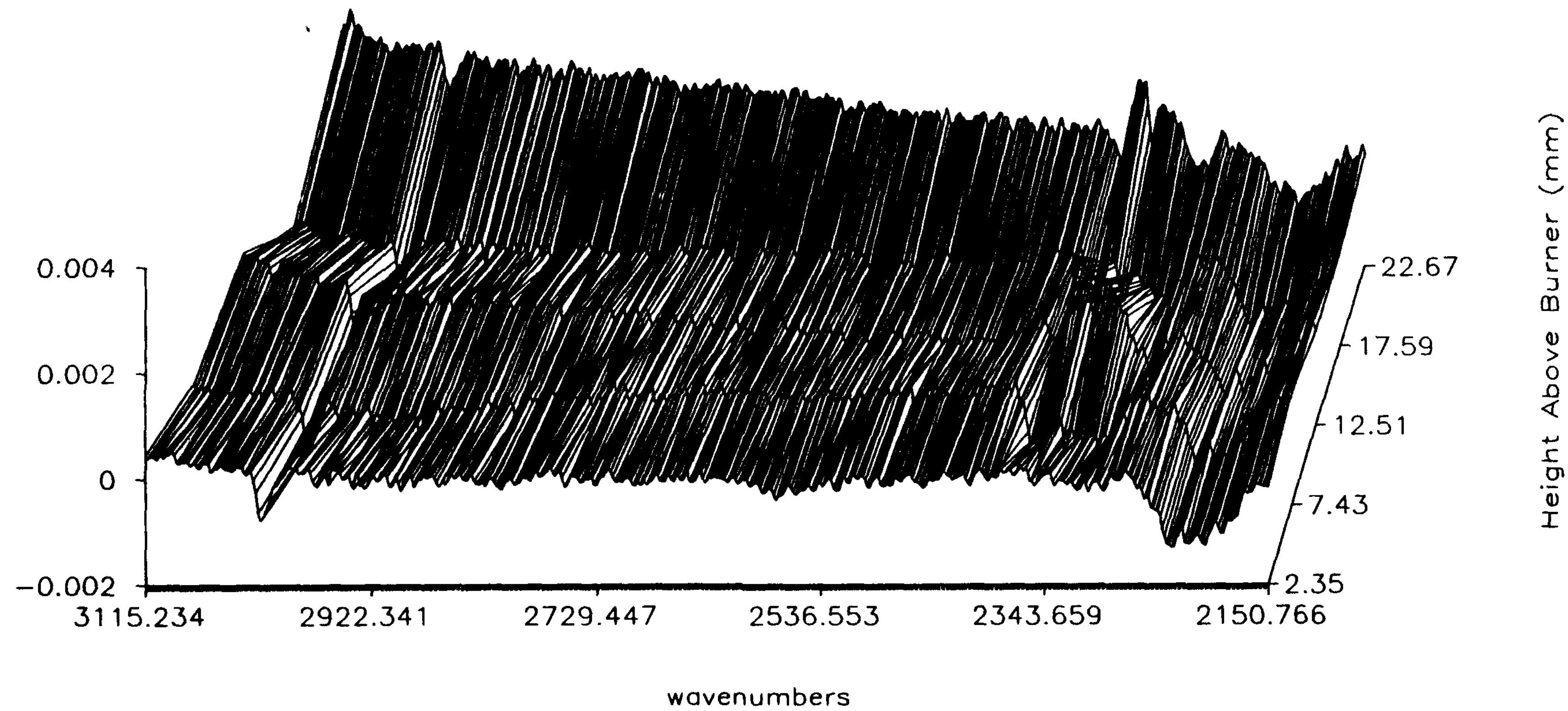

Figure 10. Infrared Spectra at the Burner Center (r = 0) for the Flowing $Ar/CH_4/N_2O$ Mixture as a Function of Height Above the Burner Surface.

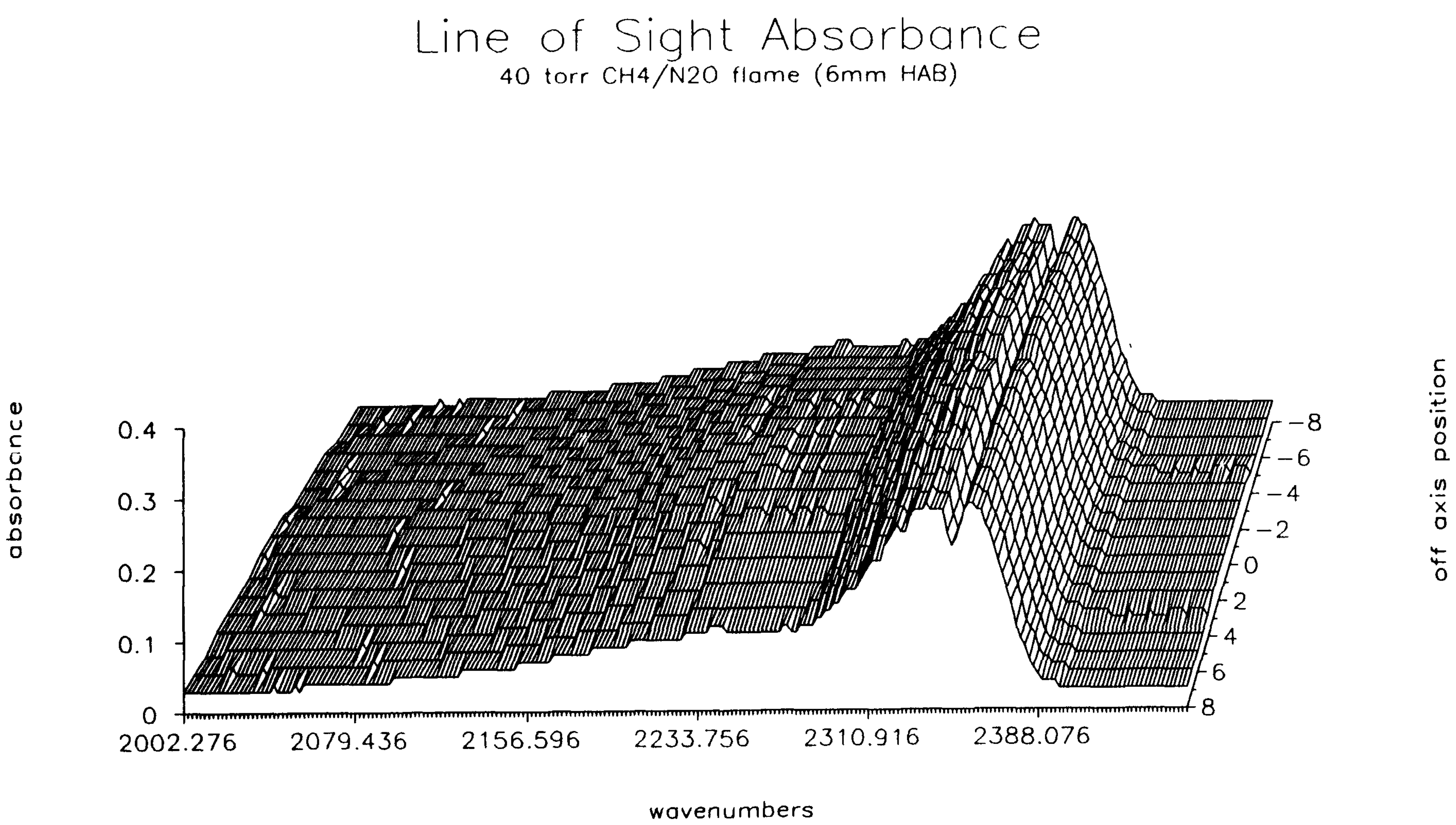

Figure 11. Line-of-Sight Absorption Spectra Through a 40-torr Stoichiometric CH_4/N_2O Flame. Data Has Been Reflected About the Burner Axis. Off-Axis Position in Units of 3 mm.

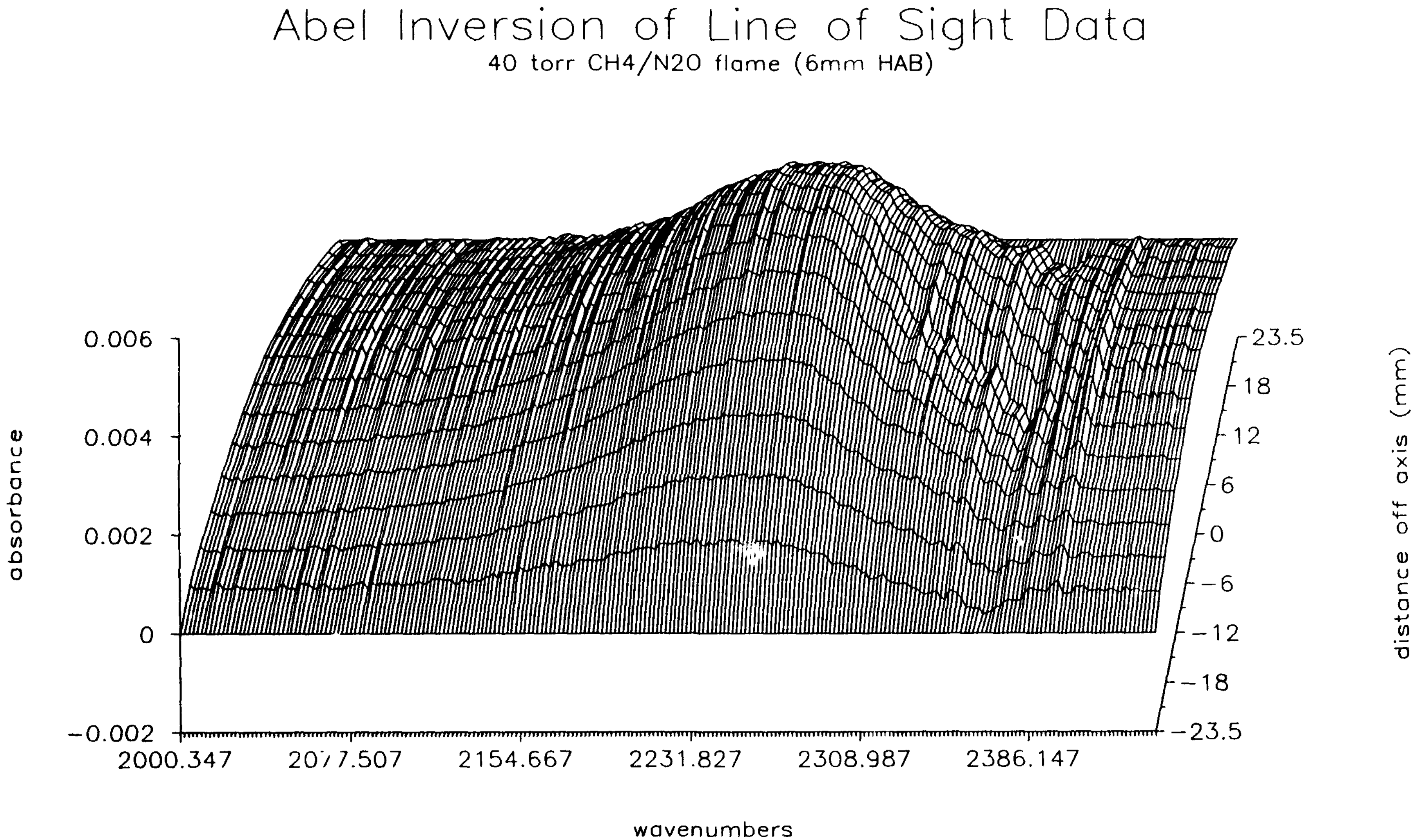

Figure 12. Inverted Data From Figure 11 Showing Almost Complete Discrimination of Absorbance Due to CO_2. Data Has Been Reflected About the Burner Axis.

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